

The University of Chicago

EXPERIMENTS IN SEARCH
OF AN EFFECT OF CENTRIFUGAL
FIELDS ON RADIOACTIVE
PROCESSES

A DISSERTATION SUBMITTED TO THE FACULTY
OF THE DIVISION OF THE PHYSICAL SCIENCES
IN CANDIDACY FOR THE DEGREE OF DOCTOR
OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY
DECEMBER, 1939

By
MELVIN LEROY SCHULTZ

CHICAGO, ILLINOIS
1945

The University of Chicago

EXPERIMENTS IN SEARCH
OF AN EFFECT OF CENTRIFUGAL
FIELDS ON RADIOACTIVE
PROCESSES

A DISSERTATION SUBMITTED TO THE FACULTY
OF THE DIVISION OF THE PHYSICAL SCIENCES
IN CANDIDACY FOR THE DEGREE OF DOCTOR
OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY
DECEMBER, 1939



By

MELVIN LEROY SCHULTZ

CHICAGO, ILLINOIS

1945



ACKNOWLEDGMENT

The author wishes to express his appreciation to Dr. Simon Freed for constant advice and encouragement during the course of this work. The author wishes also to express thanks to Dr. J. B. Hoag of the Department of Physics, to Dr. E. O. Wollan of the Chicago Tumor Institute, and to the members of the University of Chicago cyclotron staff for coöperation in making available sources of radioactive material. To Mr. Arthur Jaffey are due special thanks for much valuable assistance.



Digitized by the Internet Archive
in 2026

https://archive.org/details/IA41533905_0021

INTRODUCTION

The decay of a single radioactive substance follows the simple exponential law¹ $N_t = N_0 e^{-\lambda t}$ in which N_t is the number of radioactive atoms remaining at time t , N_0 is the number existing at $t = 0$, and λ is a constant characteristic of the substance. If λ , the decay constant, is interpreted as being the probability per unit time that a given radioactive atom will disintegrate this law may be derived from probability considerations.² It is necessary to assume that the probability of disintegration is the same for all atoms of a given radioactive isotope. However, no assumption as to the mechanism of radioactive decay need be made.

Many experiments have been performed in the search for a dependence of λ upon the external conditions to which the decaying material might be subjected.³ It has been found that the rate of decay is independent of temperature between -255°C and about 1300°C , of hydrostatic pressure up to 20,000 atm.,⁴ of the presence of magnetic fields up to 83,000 gauss, of the age of the decaying atoms, of the presence of other atoms, and of centrifugal fields of force equivalent to 20,000 times the earth's gravitational field.

A thermodynamic discussion formulated by Donnan,⁵ based on the mass energy equivalence of relativity, leads to the prediction

¹E. Rutherford, and F. Soddy, Phil. Mag. (6), 4, 370 (1902), I. Curie and F. Joliot, Compt. rend., 198, 559 (1934) have shown that artificially radioactive substances decay according to the same law.

²E. v. Schweidler, "Über Schwankungen der radioaktiven Umwandlung," Premier Congrès International pour l'étude de la Radiologie et de l'ionisation (Bruxelles: Imprimerie Scientifique L. Severeyns, 1905), pp. 1-3. K. W. F. Kohlrausch, Wien. Ber., 115, 673 (1906).

³K. W. F. Kohlrausch, Radioaktivität, Vol. XV of Handbuch der Experimentalphysik, ed. W. Wien, and F. Harms (26 vols.; Leipzig: Akademische Verlagsgesellschaft M. B. H. 1926-1937), pp. 669-676 contains a complete summary of work published prior to 1928.

⁴T. C. Poulter, Iowa Acad. Sci. Proc., 37, 311 (1930).

⁵F. G. Donnan, Nature, 104, 392 (1919).

of a change in the energy E of radioactive decay with change in gravitational potential of amount

$$\frac{\Delta E}{E} = \frac{\Delta Z}{c^2}$$

in which ΔZ is the change of gravitational potential responsible for the change in energy ΔE , and c is the velocity of light. The equivalence, on the basis of general relativity, of a gravitational and an accelerational field makes available, through the medium of the high speed air driven turbine,¹ gravitational fields whose intensities are high, up to several million g , compared with the earth's field. The difference of potential between the center and periphery of such a turbine is about 10^9 ergs/gm. The expected fractional change in energy of radioactive decay due to this difference of potential is, according to Donnan's equation, about 10^{-12} ; far outside the limits of detectability. This argument, as Compton² has already pointed out, does not exclude the possibility of the existence of a measurable effect of a gravitational field on the rate of radioactive decay. The experiments of Rutherford and Compton³ and Compton⁴ have shown that a centrifugal field of 20,000 g did not cause a change in the intensity of the gamma radiation from a radon bulb by as much as 0.1 per cent.

The performance of experiments of this general type under the more favorable conditions now possible is important, especially in view of the many new features disclosed during the rapid development of nuclear physics in the past few years. Among these features may be mentioned the discoveries of the neutron,⁵ of artificial radioactivity throughout the periodic system,⁶ of nuclear quadrupole moments,⁷ of the positron,⁸ of nuclear isomers⁹ and of

¹E. Henriot, and E. Huguenard, Compt. rend., 180, 1389 (1925). J. de phys. et rad. (6), 8, 433 (1927).

²A. H. Compton, Phil. Mag. (6), 39, 659 (1920).

³E. Rutherford, and A. H. Compton, Nature, 104, 412 (1919).

⁴Compton, op. cit.

⁵J. Chadwick, Proc. Roy. Soc., Ser. A, 136, 692 (1932).

⁶Curie and Joliot, op. cit. E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti, and E. Segrè, Proc. Roy. Soc., Ser. A, 146, 483 (1934).

⁷H. Schuler, and Th. Schmidt, Z. Physik, 94, 457 (1935).

⁸C. D. Anderson, Phys. Rev., 43, 491 (1933).

⁹W. Bothe, and W. Gentner, Z. Physik, 106, 236 (1937).

nuclear fission.¹

No detailed prediction, on the basis of the present quantum theory of nuclear phenomena, of a possible effect of the sort here considered has been made since there is not yet a definitive theory which describes both quantum and gravitational phenomena.

A simple calculation shows that the difference of potential energy, due to the centrifugal field, between an atomic nucleus on the axis of rotation of the turbine and a similar nucleus on the periphery is of the order of magnitude of 0.03 electron volts for a centrifugal field of about a million times gravity. This is far smaller than the energies, of the order of magnitude of a few million electron volts, involved in radioactive transformations. However, there are many nuclear reactions² caused by neutrons with energies of a few hundredths of a volt. Many of the product nuclei formed in these reactions are radioactive and subsequently undergo decay. A promising type of experiment then, would be one involving measurement of some of the properties of processes involving thermal neutrons. From an experimental point of view it is simpler to measure rates of decay, than quantities such as excitation functions, energies of particles, or reaction yields. In this preliminary search for an effect decay rates of naturally and artificially radioactive substances, rather than other properties, were measured.

Beta radioactive decay, especially, presents features which suggest the possibility of a gravitational effect. According to the theory of Fermi³ the process of beta decay involves transitions, within the nucleus, between neutron and proton states of the fundamental heavy particle. The transition from a neutron state to a proton state results in the emission of a negative electron and, in order that energy and nuclear spin be conserved, a neutrino. The reverse transition, from a proton state to a neutron state, is accompanied by the emission of a positron and an antineutrino. The mean lives of the beta radioactive nuclei, then, depend upon the probabilities of these transitions. The interpretation of scattering experiments involving free neutrons and protons⁴ leads to the

¹O. Hahn, and F. Strassmann, Naturw. 27, 11 (1939).

²E. Amaldi, O. D'Agostino, E. Fermi, B. Pontecorvo, F. Rasetti, and E. Segrè, Proc.Roy.Soc., Ser. A, 149, 522 (1935).

³E. Fermi, Z. Physik, 88, 161 (1934).

⁴G. Breit, E. U. Condon, and R. D. Present, Phys. Rev., 50,

conclusion that neutron-proton, proton-proton, and neutron-neutron forces are approximately equal, except for the additional Coulomb electrostatic repulsion between protons. These forces, then, appear to depend upon some property of the particles other than electrical charge. It is conceivable, therefore, that the probabilities of the neutron-proton transitions within the nucleus may be measurably altered in a gravitational field.

Independently, however, of the theory which is to be used to describe radioactive decay the possibility of obtaining intense centrifugal fields makes it imperative that effects, not only of the sort here considered, but also effects on as wide a variety of physical and chemical systems as possible, be sought.

EXPERIMENTAL

The performance of the type of experiment here described involves the spinning of a rotor containing a radioactive substance near an instrument capable of giving a quantitative measure of the intensity of the radiation from the material as a function of the time.

Because of the relatively low intensity of the neutron sources available at the start of these experiments a Geiger-Müller tube counter,¹ of a somewhat modified design, was chosen as the measuring instrument. In order that it subtend a large solid angle at the cylindrical source, a toroidal shaped counter was made. The design adopted is shown in vertical diametral cross section in Figure 1A.

The counter case, serving as the negative electrode, was made of two rings of copper A into each of which was machined a semi-toroidal groove so as to form, when the two halves were screwed together, a toroidal shaped cavity within the block. The window which admitted the beta rays to the counting region was made of a copper cylinder about 0.05 mm. in thickness and fitted into the counter as shown at B. The slight distortion of the circular cross section of the counter due to the window seemed to have no detrimental effect on the counter behavior.

The central electrode was made of the correct length of

825 (1936). B. Cassen, and E. U. Condon, Phys. Rev., 50, 846 (1936). G. Breit, and E. Feenberg, Phys. Rev., 50, 850 (1936).

¹H. Geiger, and W. Müller, Phys. Zeit., 29, 839 (1928).

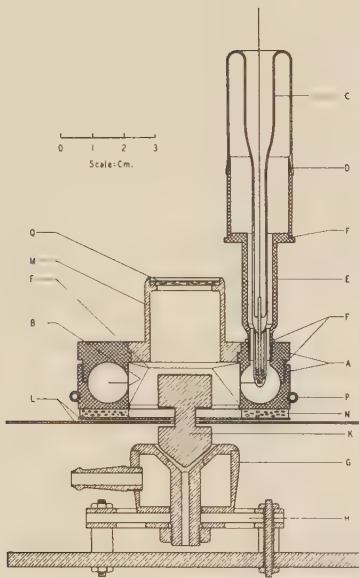


FIGURE 1A

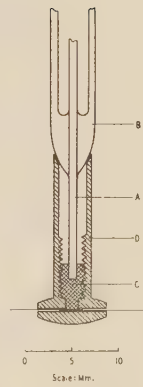


FIGURE 1B

5 mil tungsten wire bent into a circle of such diameter as to lie along the center of the toroidal cavity. It was held in position as shown in section in Figure 1B. Here A is a tungsten rod sealed through the glass support B. Silver soldered to the end of the tungsten is a threaded copper plug C which screws into the threaded lavite¹ piece D. The ends of the fine tungsten wire are brought into the lavite piece through Number 30 holes and terminate inside. The wire is held firmly in position by pressure of the copper plug. The joints between the lavite and the glass support, as well as the holes through which the tungsten wire entered, were sealed with a very small quantity of Insa-Lute²--a refractory cement.

The glass insulator, C of Figure 1A, carrying the central electrode was supported by a glass-copper seal D which, in turn, was fastened to the counter case by means of the copper support E. This insulating support was introduced into the counter through a 0.25 in. hole in the copper case and had to be put into position before the fine tungsten wire was attached.

In the most satisfactory design, the several joints F of the counter were made vacuum tight by soldering with pure tin. This type of seal permitted the baking of the counter at a moderately high temperature during its evacuation. Before the counter was assembled, the copper parts were cleaned in 6N nitric acid. After assembly, the inside was cleaned with 2N nitric acid and was subsequently rinsed several times with distilled water. The counter was then evacuated for several hours at a pressure of about 10^{-5} to 10^{-6} mm. of Hg while being baked at about 175°C . The subsequent treatment was very similar to that described by Duffendack, Lifschutz and Slawsky³ and involved the passage of an AC discharge through the counter filled with hydrogen at a pressure of a few millimeters of mercury. Alternate sputtering and pumping was continued until, with the tube containing hydrogen at a pressure of about 10 cm. of Hg, the DC voltage below which a corona discharge was no longer maintained was definite and reproducible, and until the corona current at this point was fairly large. The minimum

¹Lavite is a soapstone which is machinable. After being fired at about 1100°C . it becomes very hard.

²Manufactured by Technical Products Co., Pittsburg, Pa.

³O. S. Duffendack, H. Lifschutz, and M. M. Slawsky, Phys. Rev., 52, 1231 (1937).

current under these conditions was about 1.1 microamperes. For such a counter, the threshold lay at about 1000 volts, and the plateau was about 300-400 volts in extent. Tank hydrogen, without any purification, was found to be entirely satisfactory for use.

Some of the data here reported were obtained with counters, similar in design, but made of brass with the joints sealed by Apiezon Wax W.¹ These counters were filled with air and were not subjected to the sputtering treatment. The behavior of different counters of this type varied between wide limits. Some were nearly as satisfactory as the copper counter while some possessed no counting properties at all. Of course, no data were collected with these counters unless extended tests showed them to be reliable.

By using the circuit of Neher and Pickering² to quench the counter discharge it became possible to maintain the counter case at ground potential. The output of this quenching circuit was amplified by a single stage employing a type 57 tube and was then fed into a twofold scaling circuit similar to the first stage of the scale-of-eight circuit of Shepherd and Haxby.³ The impulses were finally recorded on a mechanical counter.⁴

The high voltage for the counter tube was supplied by a half wave rectifier the output of which was stabilized by a circuit similar to the first design of Street and Johnson⁵ employing, however, a type 57 pentode rather than a tetrode. The voltage output of the power supply was constant to within 0.2-0.3 per cent. However, the voltage actually applied to the counter depends also on the constancy of the plate current through the quenching tube of the Neher-Pickering circuit. This current; as measured by the voltage drop across a known part, at the ground end, of the cathode resistor; was usually constant to within 0.5-0.6 per cent.

The counting rate-voltage curves for a given counter and

¹Distributed by the James G. Biddle Co., 1211-13 Arch St. Philadelphia, Pa.

²H. V. Neher, and W. H. Pickering, Phys. Rev., 53, 317 (1938).

³W. G. Shepherd, and R. O. Haxby, Rev. Sci. Instr., 7, 425 (1936).

⁴Cenco high voltage impulse counter Cat. No. F791, Manufactured by Central Scientific Co., Chicago, Ill.

⁵J. C. Street, and T. H. Johnson, J. Frank. Inst., 214, 155 (1932).

radioactive source are different for different values of the grid resistor in the quenching circuit. The values used ranged from 5 to 100 megohms. Typical curves for a soldered copper counter are shown in Figure 2 for two values of the resistor, 10 and 100 megohms. Whenever the resistor is too large, a maximum appears in the curve at the start of the plateau. This is illustrated by the 100 megohm curve. The vertical line through each experimental point represents the standard deviation which is given by $n^{1/2}/t$ in which n is the total number of counts recorded for the point, and t is the length of the counting time in minutes. In general, for a constant source, the counting rate increases and the length of the plateau decreases with decreasing grid resistor. For different counters of the type here described the basic count varied from about 10 to 30 per minute, the value depending mainly on the size of the counter.

Corrections for the counting losses due to the finite resolving times of the circuits and mechanical recorded employed were made by means of multiple addition calibrations¹ in which a number of radioactive sources, each sufficiently weak to be measurable without appreciable loss, were measured singly and in combinations. The differences between the sums of the intensities of the single sources and the intensities of the same sources measured together give the losses at the several counting rates.² The calibration curves were found to be satisfactorily fitted by formulae of the Volz³-Schiff⁴ type $n = n_0 e^{-n_0 \tau}$ in which n is the observed counting rate, n_0 is the true counting rate, and τ is the overall resolving time the value of which must be determined empirically. Lifschutz and Duffendack⁵ have shown that although such a formula does not adequately represent the data when the losses become very high, it provides a satisfactory correction up to the maximum counting rate. Typical calibration curves are shown in Figure 3.

¹Duffendack, Lifschutz, and Slawsky, op. cit. H. Lifschutz and O. S. Duffendack, Phys. Rev., **54**, 714 (1938).

²By the use of dummy source holders the geometrical arrangement was the same for each measurement making the fraction of the radiation scattered into the counter always the same.

³H. Volz, Z. Physik, **93**, 539 (1935).

⁴L. I. Schiff, Phys. Rev., **50**, 88 (1936).

⁵Lifschutz and Duffendack, op. cit.

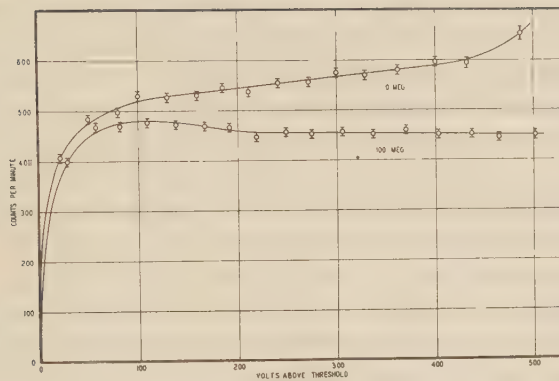


FIGURE 2

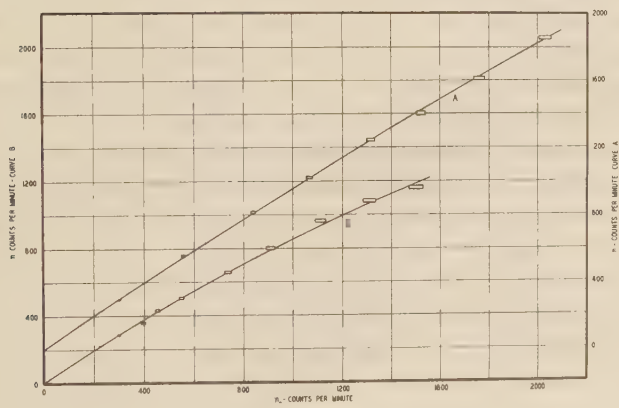


FIGURE 3

Curve A refers to a soldered copper counter with a 10 megohm grid resistor and B to a wax sealed brass counter with a 100 megohm resistor. The values of τ for the two curves are 0.00281 sec. and 0.00948 sec. respectively. The rectangles represent the magnitudes of the standard deviations of the experimental points.

The rotors used were of the general type first described by Henriot and Huguenard¹ and developed by Beams and his co-workers.² The dimensions and details of construction of the rotors used will be included in the discussion of the results of the individual experiments. It was found that the maximum speed of a given rotor depended very markedly on the angle of the cone; becoming a maximum for the most acute angle, about 102° , permitting stable rotation. The turbine blades were simply radial grooves cut with a 60° end mill; they ranged in number from 24 to 60 and in depth from 0.005 to 0.020 in. Within limits, the rotational speed increased with increasing depth of the grooves. The maximum speeds obtainable from rotors of the type used were from about 4,000 to 5,000 rev./sec. with driving air pressures up to 90 lbs./sq. in.

The stator, illustrated by G of Figure 1A, was made of brass and was similar in design to the one described by Beams. The cone angle was 92° . The jets, twelve Number 73 holes emerging on a circle of radius 0.56 cm., made an angle of 30° with the horizontal. The horizontal projection of the jets made an angle of 8° with a tangent to the cone. The stator was mounted on a brass plate by means of three legs fastened to the gum rubber washer H. The orientation of the stator was maintained by a clamp around the rubber pressure tubing admitting the compressed air.

The stator assembly was mounted on a portion of the slide rest from a jeweler's lathe in order to provide a means of centering the rotor within the counter. This centering was rather critical if the diameter of the rotor was not very much less than that of the counter window. This complete assembly was mounted on a lead plate 30 cm. in diameter and 3 cm. thick which, in turn, was mounted on a vertical way to permit the raising and lowering of the rotor into and out of the counter which was fixed below a similar lead plate 7 cm. thick. To enable the rotor to be reproducibly

¹Henriot, and Huguenard, op. cit.

²J. W. Beams, and E. G. Pickels, Rev. Sci. Instr., 6, 299 (1935).

replaced within the counter, the slide of the way was provided with a scale and vernier which could be read to 0.005 cm.

Unless special precautions were taken radium and thorium active deposit, always present in the atmosphere, collected on the counter and surrounding objects from the compressed air used for driving the rotor.¹ The radiation from this material, up to about fifty counts per minute in intensity, proved to be a serious interference with any attempted measurements. By constructing the rotor as shown at K in Figure 1A it became possible to use a system of two baffle plates L, with a small gap between, to shield the counter from most of the driving air. The baffle plates were made in two halves so that they could be withdrawn during the starting and stopping of the rotor or be put into place once the rotor had attained speed. By further closing the space around the upper part of the rotor by means of a cap M over the counter and a one-eighth inch thick sponge rubber washer N between the counter and the upper baffle it became possible to reduce the magnitude of this air effect until it was usually not more than five to ten counts per minute above the basic rate.² To prevent an excessive temperature rise in the counter due to the spinning of the rotor in the confined space the counter was provided with a single turn P of one-eighth inch copper tubing through which water was circulated.

The angular velocities of the rotors were measured by means

¹Some of the properties of these deposits and of the process of deposition were studied in considerable detail. Since the results are to be recorded in detail elsewhere only a brief summary will be given here. Measurements of the decay rates of a large number of these deposits show definitely that they consist of radium and thorium active deposits. Some specificity in the deposition of the material on various substances was found. The material seemed not to deposit on lead surfaces at all. Some tank gases, namely electrolytic oxygen and oxygen prepared from liquid air were strongly active while others, nitrogen, hydrogen, and carbon dioxide were inactive. The amounts of material obtained from oxygen aged for times long compared with the half life of radon cannot be explained unless the radium contamination of the steel tank is assumed to be abnormally high. The passage of air through a charged cylindrical condenser indicates that the radioactive material which deposits is unchanged.

²The intensity could still further be reduced by passing an inactive gas such as carbon dioxide into the space around the upper part of the rotor. In spite of the lower viscosity of carbon dioxide this resulted in a reduction in speed of the rotor of about 250 cycles when the total pressure inside was about 5 mm. of Hg above atmospheric; for this reason the method was usually not employed.

of a stroboscope consisting of a small 110 volt neon bulb driven by the amplified output of a beat frequency oscillator whose calibration was guaranteed to two per cent by the manufacturer.¹ The light from one electrode of the bulb was reflected through the glass window Q, in the cover over the counter, to the upper surface of the rotor, on which was painted a black spot, and from the rotor through the window to a second mirror which enabled an observer to view the top of the rotor from a horizontal direction. No special precautions were taken to keep a constant rotational speed, but usually the maximum variation was less than 100 rev./sec. even over many hours.

In order to determine whether or not the response of the counter was materially altered by the presence of the spinning rotor, the gamma rays from a 2 mg. radium source were measured at several standard positions along a graduated steel rod while a rotor was spinning within the counter and also in the absence of a rotor. The average ratio of the counting rate with the rotor spinning to the counting rate in the absence of the rotor was found to be 0.973 with an average deviation of 0.017 over a counting range up to about one thousand per minute. Since the source was at the same height as the counter the slightly lower rate with the rotor spinning is probably due to the shielding effect of the rotor on the part of the counter away from the source.

The actual measurements of the decay rates were carried out according to the following procedure. The rotor containing the activated material was set into rotation while the stator was in the completely lowered position. After the maximum speed had been attained, the two halves of the system of baffle plates were clamped in place around the rotor. Then, the whole assembly was raised into position until the gap between the sponge rubber washer N and the upper baffle plate was closed. From this point on, the measurement consisted of recording the number of impulses passing through the counter during definite intervals of time measured by means of a stop watch; the lengths of the intervals depended upon the rate of decay of the source and upon its intensity.

Because it was impossible to eliminate completely the effect of the radioactive material in the compressed air it became necessary to correct the normal basic count for this added effect.

¹The Clough-Brengle Co., 5501 N. Broadway Avenue., Chicago, Ill.

Since the material deposited is complex, consisting of the short lived radium active deposit and the long lived thorium active deposit in varying amounts depending upon a variety of meteorological conditions, the rate of rise of the excess activity appeared not to follow exactly any simple law. From the study of a large number of cases of the rise in rate while unactivated rotors were spinning it was found that over the course of a few hours the rise could approximately be represented by a linear function of the time. In the measurement of sources with short periods, then, the correction was made by measuring the basic count before the start of the experiment and again after the source had decayed and assuming a linear change during the course of the run.

For long period decays, when the rotor was required to spin continuously for 10 to 20 hours or more, such a method of correction was not reliable. In these cases, at intervals of a few hours, the rotor, while spinning at constant speed, was completely lowered from the counter. Between the rotor and the counter was placed a lead disc 16 cm. in diameter and 2.5 cm. thick to screen the latter from the beta and gamma rays emitted by the source. The counting rate with this arrangement was assumed to be due to the normal basic count and to the active deposit on the counter. A small correction had to be applied to this observation for two reasons. With the stator assembly lowered: (1) the shielding of the counter from stray radiation is less effective, and (2) the radiation from the active deposit on the parts of the stator assembly is not counted while with the stator raised into position it is to some extent. These two effects are in opposite directions and nearly compensate. The correction factor, the ratio of the counting rate with the stator in position to the rate with the stator lowered, was determined using unactivated spinning rotors. From twelve determinations it was found to be 0.97 with an average deviation of 0.03. The counting rates, corrected for these effects, were plotted as a function of the time so that the rate for any given time could be read from the smooth curve drawn through the points.

The method of deduction of the decay rates from the data obtained in the manner described above differed depending upon whether the observed data could be represented by a single exponential period or whether the sum of two or more exponentials was required. For the former case, the decay constants were computed

by Peierls' method¹ which requires only the observation of the numbers of counts during equal intervals of time, in length, less than $0.3/\lambda$ where λ is the decay constant.

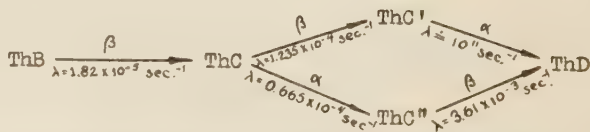
This method is not applicable if the decay cannot be represented by a single exponential. In the latter case the observed counting rate N may be written as a sum of exponential periods in

$$N = \sum_1 N_{01} e^{-\lambda_1 t}$$

which 1 is the number of periods required to represent the decay curve. The value of the initial intensity N_{01} , for each period, was obtained from the experimental data by the method of least squares assuming the literature values for the λ_i to be correct. In order to avoid the necessity for employing weighting factors the length of observation time for each experimental point was so chosen as to make the relative probable error for that point approximately equal to the errors of all the other points. If any systematic deviation between the curve, calculated from the N_{01} and the λ_1 , and the experimental points is significantly larger than the experimental error it may be concluded that at least one of the decay constants involved is significantly different from the assumed value.²

RESULTS

Thorium active deposit.--The first experiments were carried out with thorium active deposit consisting of ThB, ThC, ThC', and ThC'' in transient equilibrium. These substances are related in the following manner.



The sources were prepared by collecting on the outside upper cylin-

¹R. Peierls, Proc. Roy. Soc., Ser. A, 149, 467 (1935).

²It may be shown very simply that it is impossible for any changes in the values of the λ_i , larger than the experimental error, to occur without being revealed by this method of treatment of the data so long as the decay in the presence of the centrifugal field remains expressible as a sum of exponentials.

dric surface of the steel rotor shown in section in Figure 4A, charged negatively to about 300 volts, the positively charged ThA atoms formed by the decay of the thoron from about 400 gms. of thorium nitrate. The decay of such a source should show only a single exponential period with the 10.6 hr. half life of ThB.¹ For none of the four sources of this type, measured at speeds of about 3800 rev./sec., was the decay completely exponential. The shapes of the curves were not reproducible. Some of them showed exponential decay with approximately a 10.6 hr. period during part of the measurement but some of them did not.

In an attempt to localize the source of these abnormalities the next experiments were performed using ThC, in equilibrium with ThC' and ThC'', which had been separated from ThB by repeated coprecipitation with ferric fumarate according to the method of McCoy and Viol.² This type of source should normally decay exponentially with the 60.8 min. half life characteristic of ThC.³ The thorium active deposit used in these separations was collected on a negatively charged platinum foil exposed to thoron from a radiothorium preparation.⁴

The separated ThC was deposited, by replacement from a hydrochloric acid solution, on the inside surface of the thin wall of the steel rotor illustrated in Figure 4B. The rotor wall was 0.36 mm. thick at the top tapering to 0.70 mm. at the bottom over a height of 4 mm. The deposition of ThC was confined to this surface by a coat of lacquer over the remaining surfaces.

With these sources, as well, deviations from exponential decay were observed. A typical curve is shown in Figure 5 where the counting rate, on a logarithmic scale is plotted against the time. The vertical lines represent the standard deviations of the individual points. The linear portions of the decay give half lives which are, within experimental error, equal to the published value. The general shape of the portions of the curve showing

¹F. v. Lerch, Wien. Ber., 116, 1443 (1907).

²H. N. McCoy, and C. H. Viol, Phil. Mag. (6), 25, 353 (1913). By the addition of a small amount of an inactive lead salt, isotopic with ThB, the separation was effected in two or three precipitations.

³Ibid.

⁴This material was kindly lent by Dr. J. B. Hoag of the Department of Physics.

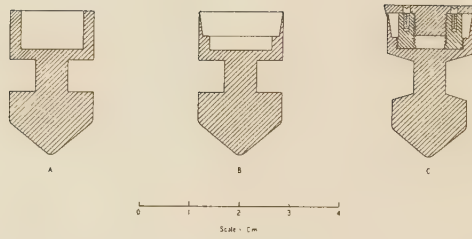


FIGURE 4

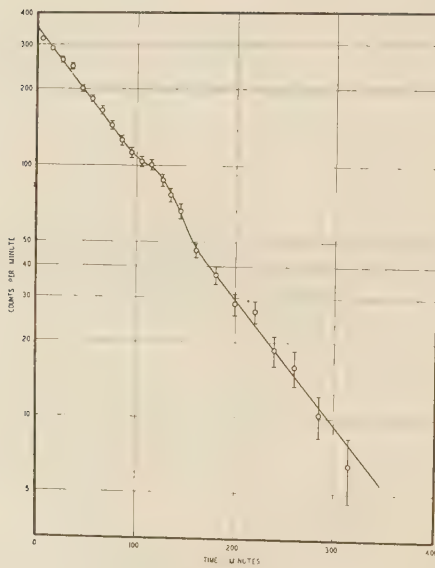


FIGURE 5

deviations from exponential decay was reproducible from experiment to experiment but the point along the curve at which the deviation appeared was not.

When the experiment was performed with the covered rotor of Figure 4C, the material again having been deposited on the inside surface of the thin wall, no such deviations appeared; the decay was exponential throughout the whole measurement. Similarly, when the open rotor was used with a film of lacquer covering the source no abnormalities were evident. From this, one may conclude that the irregularities observed were due probably to loss of material either mechanically or by aggregate recoil as a result of the alpha decay of ThC and ThC'. It is rather difficult to understand, however, why such transport of material should occur during only a small fraction of the total decay.

The results of the measurement of the half lives of six covered sources are shown in the Table together with the results of some of the other experiments to be discussed. The errors quoted for the observed half lives are those obtained by Peierl's method; they include only the effect of statistical fluctuations. The column headed air effect gives the magnitude of the rise, in counts per minute, above the normal basic count due to the radioactive material in the driving air. The column labeled n gives the speed in revolutions per second, of the rotor used; R is the average radius of the ring of radioactive material. The centrifugal field strengths, expressed as multiples of the earth's gravitational field, were calculated from the formula

$$\frac{4\pi^2 n^2 R}{980}$$

in which 980 is approximately the gravitational acceleration of the earth. These values are tabulated in the last column. The weighted average of the half lives found for ThC is 59.4 min. with an average deviation of 1.4 min. Within this limit, the half life found does not differ from the published value. The rather large deviations from the average are undoubtedly due, in part at least, to uncertainties introduced by the correction for the air effect.

Manganese.--Radioactive Mn^{56} , which may be formed by the reaction $\text{Mn}^{55}(n,\gamma)\text{Mn}^{56}$, with a half life of 2.59 ± 0.02 hrs.¹ has a beta ray spectrum showing two distinct groups of electrons with

¹J. J. Livingood, and G. T. Seaborg, Phys. Rev., 54, 391 (1938).

SUBSTANCES SHOWING SINGLE DECAY PERIODS

Radioactive Isotope	Literature Half Life	Observed Half Life	Air Effect Counts/min.	Initial Intensity Counts/min.	Final Intensity Counts/min.	R Cm.	n rev./sec.	Centrifugal Field $g \times 10^{-5}$
ThC	60.8 min.	57.6 $\pm$ 0.3 min.	15	616	12	0.84	3500	4.15
ThC	60.3 min.	62.4 $\pm$ 0.4 min.	0	614	17	0.79	4000	5.09
ThC	60.3 min.	58.0 $\pm$ 0.6 min.	7	353	13	0.79	3950	4.97
ThC	60.8 min.	59.2 $\pm$ 1.0 min.	1	143	9	0.79	3950	4.97
ThC	60.8 min.	61.2 $\pm$ 1.6 min.	9	316	126	0.79	3900	4.84
ThC	60.8 min.	59.2 $\pm$ 2.3 min.	0	50	5	0.79	3900	4.84
Mn ⁵⁶	2.59 $\pm$ 0.02 hr.	2.65 $\pm$ 0.03 hr.	0	172	29	0.79	4025	5.16
Mn ⁵⁶	2.59 $\pm$ 0.02 hr.	2.55 $\pm$ 0.08 hr.	0	60	17	0.85	4000	5.48
Cl ³⁸	37-40 min.*	39.1 $\pm$ 0.2 min.	15	1489	43	0.79	4025	5.16
Cn ⁶⁴	12.8 $\pm$ 0.1 hr.	13.01 $\pm$ 0.02 hr.	5	1420	224	0.85	3700	4.69

*See text for discussion of discrepancies in the half lives reported for Cl³⁸.

upper limits at 1.2 and 2.9 million electron volts.¹ This indicates that the Fe⁵⁶ nucleus resulting from the decay of Mn⁵⁶ is left sometimes in an excited state and sometimes in the ground state. Within rough limits, Brown and Mitchell have found that the two groups decay with the same half life.

Finely divided MnO₂ was irradiated for about thirty hours with slow neutrons from a 200 millicurie radon-beryllium source contained in a small soft glass bulb which was placed at the center of a paraffin cylinder 23 cm. in diameter and 36 cm. high. The MnO₂, also embedded in the paraffin, was separated from the source by about 6 cm. After irradiation, the activated material was placed in the cavity of the thin-walled, covered rotor shown in Figure 4C. The wall thickness of this rotor was approximately the same as that of the rotor of Figure 4B. The escape of material

¹M. V. Brown, and A. C. G. Mitchell, Phys. Rev., 50, 593 (1936).

from the cavity was prevented by a paper gasket between the rotor and cover and by the overhanging edge of the cover. The unscrewing of the cover during rotation was prevented by two 0-80 hardened steel set screws passing into the body of the rotor through holes in the cover. During the spinning the material packs tightly against the outer wall of the rotor. This redistribution is completed during the first few minutes and does not, after that, influence the shape of the decay curve. The decay was found to be exponential with a period of 2.65 ± 0.03 hrs. as evaluated by Peierls' method. This value, within experimental error, agrees with the published value. The details of the speed and centrifugal field are given in the table.

Iron.--Iron irradiated with fast neutrons gives the same radioactive manganese isotope Mn^{56} by the reaction $Fe^{56}(n,p)Mn^{56}$. The steel rotor of Figure 4B was activated by being placed as closely as possible to a 235 mc. Rn-Be neutron source.¹ The half life was found to be 2.55 ± 0.08 hrs. which differs from the published value by less than the experimental error.

Chlorine.--The beta ray spectrum of Cl^{38} similarly shows two groups² indicating that here also the product nucleus, A^{38} , may be left in either an excited state or in its ground state. There are some discrepancies among the published half lives for Cl^{38} . Van Voorhis³ has reported 37.0 min. from deuteron activated $LiCl$. Hurst and Walke⁴ found a 37.5 min. period from slow neutron activation of $PbCl_2$ and NH_4Cl . Kurie, Richardson and Paxton⁵ have reported, without giving details, that various persons working in their laboratory have found values ranging from 39 to 40.5 min.

About one gram of KCl , wrapped in 0.001 in. thick aluminum

¹Since fast neutrons are so penetrating, the whole rotor was activated. Some beta rays from parts of the rotor other than the outside upper cylindrical surface were undoubtedly counted. For this reason, the radiation measured cannot be assumed to have come entirely from a region of homogeneous centrifugal field. The value of R given in the table is the radius of the outer cylindrical surface.

²F. N. D. Kurie, J. R. Richardson, and H. C. Paxton, Phys. Rev., **49**, 368 (1936).

³S. N. Van Voorhis, Phys. Rev., **49**, 889 (1936).

⁴D. G. Hurst, and H. Walke, Phys. Rev., **51**, 1033 (1937).

⁵Kurie, Richardson, and Paxton, op. cit.

foil, was bombarded for about one minute with about ten microamperes of 5.5 MEV deuterons from the University of Chicago cyclotron. The chlorine was precipitated from the aqueous solution of the activated KCl as AgCl. A small amount of this active material was mixed with inactive AgCl and was placed inside the covered rotor of Figure 4C. The half life was found to be 39.1 ± 0.2 min. In view of the disagreement between the values reported in the literature, although the 37 min. values are probably the more reliable, the value found here cannot be said to differ significantly from the value to be expected from the decay of a stationary source.

Copper.-- Cu^{64} , formed by the reaction $\text{Cu}^{63}(\text{d},\text{p})\text{Cu}^{64}$, has a period of 12.8 ± 0.1 hrs. and decays by a branching process to Ni^{64} by positron emission and to Zn^{64} by electron emission.¹ Both types of decay have been found to show the same period.

A beryllium copper alloy² rotor of the type shown in Figure 4A was exposed for 45 sec. to about ten microamperes of 5.5 MEV deuterons in a holder which permitted access of the beam only to the upper outside cylindrical surface. The rotor was turned in the beam so as to make the activation approximately uniform over the surface. Since the range of the deuteron beam in the copper is only about 0.05 mm., the radiation measured comes from a region of homogeneous centrifugal field on the periphery of the rotor. The measurement was commenced only after all of the short periods due to contamination and to Cu^{66} had decayed. The half life was found to be 13.01 ± 0.02 hrs. The standard deviation quoted is that obtained by Peierls' method and takes into account only the error introduced by the statistical fluctuations of the numbers of particles counted but does not include the error introduced in correcting for the air effect and for the non-linearity of the response of the counter and recorder. When these are included the error is about 2 per cent. The observed period probably does not, then, differ significantly from the published value.

Aluminum.--Aluminum bombarded by neutrons displays three radioactive periods due to the reactions $\text{Al}^{27}(\text{n},\text{p})\text{Mg}^{27}$; $\text{Al}^{27}(\text{n},\alpha)\text{Na}^{24}$; and $\text{Al}^{27}(\text{n},\gamma)\text{Al}^{28}$. The half lives are 10.2 min.,³ 14.8

¹S. N. Van Voorhis, Phys. Rev., **50**, 895 (1936).

²The nominal composition is: Be-2.25%; Ni-0.50% Fe-0.10%; Cu remainder.

³M. C. Henderson, Phys. Rev., **48**, 855 (1935).

hrs.,¹ and 2.36 min.² respectively. The two latter isotopes, Na²⁴ and Al²⁸, decay always to excited states of the product nuclei, Mg²⁴ and Si²⁸, the transitions to their ground states being forbidden by selection rules.

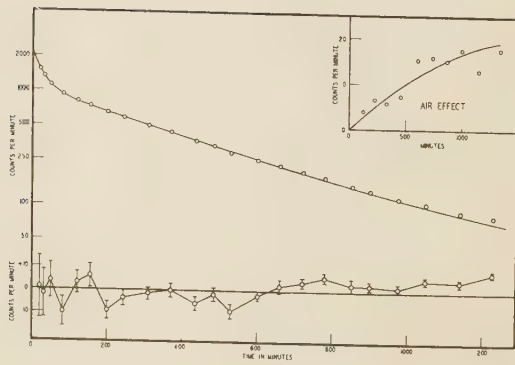
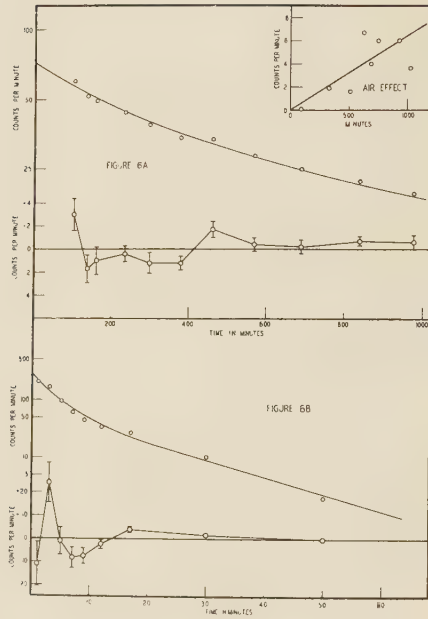
A solid duralumin³ rotor as shown at K in Figure 1A was activated by a ninety-hour exposure to the neutrons from a 160 mc. Rn-Be source embedded in the paraffin cylinder, the arrangement used being similar to that employed for manganese. The decay curve obtained was resolvable into four exponential periods which were ascribed to Mg²⁷, Na²⁴, Al²⁸, and Mn⁵⁶; the latter being produced, in measurable intensity, from the 0.5 per cent manganese in the alloy. The data were treated according to the second method discussed in the experimental section.

Because of the large difference between the half lives of the two short and the two long periods, the initial intensities for the two long periods were first obtained from the data commencing after the first one-hundred minutes of the decay. The two short periods were similarly treated separately after first subtracting the calculated intensities due to the long periods from the observed intensities. The curves so obtained are plotted in Figure 6 A and B for the long and short periods respectively. The counting rates in impulses per minute are plotted against the time in minutes. Below each decay curve are plotted, on an enlarged scale, the deviations between the calculated curve and the experimental points. The vertical lines through the points represent the standard deviations. It may be seen that the deviations are not significantly larger than those to be expected from the statistical fluctuations in the distribution of counts. These statistical fluctuations are large because the intensities were low. The initial intensities, in counts per minute, for the four periods were as follows: Al²⁸ - 207.9; Mg²⁷ - 69.0; Mn⁵⁶ - 32.1; and Na²⁴ - 39.8. The radius of the rotor was 0.853 cm.; the average speed was 3700 rev./sec. giving, on the periphery of the rotor, a field of 4.7×10^5 g. In this experiment a considerable fraction of the

¹S. N. Van Voorhis, Phys. Rev., **49**, 889 (1936).

²C. D. Ellis, and W. J. Henderson, Proc. Roy. Soc., Ser. A, **156**, 358 (1936).

³The alloy used, 17ST, has the nominal composition:
Cu - 4.0%; Mn - 0.5%; Mg - 0.5%; Al - remainder.



radiation originated in parts of the rotor where the field was smaller than this value. It may be roughly calculated that about 90 per cent of the radiation originated within 1.5 mm. of the periphery of the cylindrical part of the rotor. The inset in the upper right hand corner of Figure 6 shows the rise in basic count due to the air effect.

Bromine.--The two radioactive bromine isotopes, Br^{80} and Br^{82} , formed by deuteron or slow neutron bombardment of bromine, exhibit three decay periods;¹ 18.5 ± 0.5 min., 4.54 ± 0.10 hrs., and 33.9 ± 0.3 hrs. Br^{80} has been shown to be responsible for two of them.² The 4.5 hr. period has been shown to be due to an excited state of Br^{80} decaying by gamma ray emission to the ground state which in turn decays, by electron emission with an 18.5 min. period, to Kr^{80} .³

In the first experiment about 150 gms. of NaBrO_3 were irradiated with slow neutrons from a 185 mc. Rn-Be source. The radioactive bromine was separated by adding to the solution of the NaBrO_3 a small amount of NaBr and precipitating the bromine as AgBr . This material was placed in the covered hollow rotor as before. The decay curve obtained was resolvable graphically into three periods of approximately 40 min., 6 hrs., and 34 hrs. The value of R was about 0.79 cm.; the average speed was 3975 rev./sec.; and the centrifugal field about $4.8 \times 10^5 \text{ g}$. There is a possibility that, since the strongly activated Na^{24} was present in the NaBrO_3 in large amounts, the changes in the apparent half lives were due to some 14.8 hr. Na^{24} having been carried down during the precipitation of the AgBr . To eliminate this possibility a second experiment was performed in which LiBr was bombarded for three minutes with three to four microamperes of 8.5 MEV deuterons. The Li^8 which is formed along with the radioactive bromine, has a half life of 0.88 sec. and cannot interfere. Silver bromide was precipitated from the solution of the activated LiBr . This material was placed on the rotor in the following manner. A solid beryllium copper rotor was silver plated over its upper cylindrical surface. The

¹A. H. Snell, Phys. Rev., **52**, 1007 (1937).

²Bothe, and Gentner, op. cit.

³E. Segrè, R. S. Halford, and G. T. Seaborg, Phys. Rev., **55**, 321 (1939). D. C. DeVault, and W. F. Libby, Phys. Rev., **55**, 322 (1939).

rotor was heated in an electric furnace to a temperature just above the melting point of AgBr. While at this temperature, small amounts of the molten AgBr were transferred to the silver surface. This material wets the silver surface and forms a very adherent layer after solidification. Before being set into rotation, the layer was vigorously buffed to remove any loosely adherent material.

The decay curve shown in Figure 7 was obtained with this source. R was 0.634 cm; the speed was 4975 rev./sec.; and the centrifugal field was 6.32×10^5 g. The continuous curve was calculated from the data in the manner already discussed. The experimental points again are shown by vertical lines. An examination of the plot, on an enlarged scale, of the differences between the calculated curve and the experimental points shows a small systematic trend starting at about 200 minutes. Since the standard deviations, represented by the vertical lines, do not include the non-linearity and air effects this trend is probably within the total experimental error since, as is shown in the inset in the upper right hand corner of the figure, the air effect in this case was quite large. In the absence of any further information it must be concluded that the large changes in the apparent half lives observed in the first experiment were due to the presence of some radioactive sodium.

CONCLUSIONS AND DISCUSSION

The results obtained may be summarized by the statement that a centrifugal field of about five-hundred-thousand times the earth's gravitational field has been found not to change the radioactive decay rates of ThC , Mn^{56} , Cl^{38} , Cu^{64} , Mg^{27} , Na^{24} , Al^{28} , Br^{80} , or Br^{82} by as much as three per cent. These substances were chosen for measurement since they are representative examples of several types of decay showing special features. The features of interest in each case have been pointed out in the discussion of the individual experiments.

The precision of the measurements has been limited mainly by the following factors: (1) the necessity, because of the low intensity of radioactive sources activated by radon-beryllium neutron sources, for using a Geiger-Müller counter as the measuring instrument; (2) the corrections required because of the non-linearity of the response of the recording circuits; (3) the corrections necessary to take into account the radioactive material deposited

from the driving air.

The precision could materially be increased by changes, of the following sort, in the apparatus. By making use of the cyclotron as a means of activating sources to much higher intensities than may be obtained in other ways an ionization chamber and direct current amplifier could be used as the measuring instrument. The sensitivity of an ionization chamber remains much more constant, especially over long periods of time, than that of a counter. Because of the much higher intensity which may be used the statistical fluctuations, due to the random time distribution of the radioactive disintegrations, will be much smaller. With these higher intensities the air effect would become relatively less important. If it were found not to be negligible under these conditions it could be completely eliminated by using a rotor spinning in a vacuum chamber in the manner which has been extensively used in ultra centrifuge work.¹ The rotor in the chamber is driven from small turbine spinning in air, by means of a shaft passing through an oil gland. This method would permit, also, a much greater freedom in the shapes of the rotors which might be employed.

In conclusion it might be of some interest to point out briefly some other types of experiments which might be performed. Since the ionization chamber method gives a continuous reading it would be possible to follow changes which might occur over short intervals of time. For example, one might expect to find changes occurring during acceleration or deceleration of the spinning rotor analogous to the effects observed in electromagnetic induction where a voltage is induced only by changes in magnetic flux. Nuclear reactions in which uncharged particles are absorbed or emitted are promising for study, especially those in which slow neutrons are absorbed. For nuclei possessing quadrupole moments it may be possible to find directional effects on, for example, the yield of a reaction as a function of the relative orientation of the beam of incident particles causing the reaction and the direction of the centrifugal field vector. Nuclear fission; in which the nucleus is divided into parts of comparable mass as a result of neutron bombardment, as has been observed for thorium and uranium; presents an entirely new field of experience which would be very interesting to study from the point of view of the effect of a centrifugal field.

¹Beams, and Pickels, op. cit.

